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Effect of BET surface area of recycled U_3O_8 powder on Mn-Al doped UO_2 pellets property

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1. Introduction

Extending the fuel discharged burn-up, while enhancing the safety features is one of the major challenges to nuclear energy industries because it can reduce the maintenance and fuel cycle cost [1]. Research on fuel pellets focuses on increasing the pellet density and grain size to increase the uranium contents and the high burn-up safety margins for LWRs[2].

In the commercial manufacturing of UO_2 fuel pellets, defective UO_2 pellets that do not meet the manufacturing specifications of density and diameter may be produced. Defective UO_2 pellets or scraps of pellets should be reused in manufacturing new UO_2 pellets. A commonly used recycling method for defective UO_2 pellets or scraps of pellets is that defective UO_2 pellets are oxidized in air at about 450℃ to make U_3O_8 powder and then added to UO_2 powder[3]. Because the recycled U_3O_8 powder has a low sinter-ability compared to the raw UO_2 powder, addition of U_3O_8 to UO_2 leads to a drop in the pellet density, impeding a grain growth and the formation of undesired graph-like pore segregates. This adverse effect of recycled U_3O_8 powder on UO_2 pellet property becomes more severe in large grain sized UO_2 pellets. In general, the sinter-active powder should have small size and large BET surface area. However, since the powder property of U_3O_8 depends on the initial grain size of UO_2 pellets, larger grain UO_2 pellet produces poorer sinter-active U_3O_8 powders having larger particle size and smaller BET surface area.

In this paper, the effects of several process parameters on U_3O_8 powder properties have been tested. It is suggested that BET surface area of recycled U_3O_8 powder can be increased by applying low temperature oxidation and/or sequential cyclic thermal treatment procedures. The effect of those recycled U_3O_8 powder on the UO_2 pellet properties is also investigated. The U_3O_8 powders having higher BET surface area can mitigate the UO_2 pellet density drop.

2. Experimental

The Mn-Al doped UO_2 fuel pellets were prepared by a conventional sintering process. Recycled U_3O_8 powders were prepared by heating the Mn-Al doped sintered pellets in air at 325℃ and 450℃, respectively. In order to reduce the particle size thereby increase BET surface area further, the above two kinds of obtained U_3O_8 powers were ball-milled or cyclic heat treated, respectively. In the cyclic heat treating, the U_3O_8 powder was reduced and then re-oxidized, repetitively. The BET surface area of U_3O_8 powders were measured with a BET surface area analyzer.

5 and 10wt% of those U_3O_8 powders were added to the mixture of $MnO-Al_2O_3$ powder and UO_2 powder. Total amount of Mn-Al in the powder mixture was controlled to be 1000ppm in weight with considering the Mn-Al weight in recycled U_3O_8 powder. Powder mixtures were mixed with a tumbling mixer. The powder mixture was pressed into green pellets at 300 MPa. The green pellets were sintered at 1730℃ for 4h in flowing H_2 gas.

The sintered density of the UO_2 pellets was measured by the water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290℃ in carbon dioxide gas in order to examine their grain boundaries. The grain structures were examined by an optical microscope and the grain size was determined by the linear intercept method.

3. Results

Generally, a powder having a large BET surface area and a smaller particle size is more effective to mitigate the degradation of UO_2 pellet properties which is usually observed in recycled U_3O_8 containing UO_2 pellets. So, in this study, we have focused on the process parameters which could increase the BET surface area of U_3O_8 powder effectively.

Fig.1 shows the variations of BET surface area with the processing parameters such as the oxidizing temperatures, repetition of reducing and then re-oxidizing, and a ball milling of the oxidized powder. The BET surface area of the recycled U_3O_8 powder increased when UO_2 pellets was oxidized at relatively low temperature of 325℃. Milling of the oxidized powder seems to be an ineffective process to increase the BET surface area. The BET surface area of U_3O_8

powder oxidized at 325℃ did not changed after milling, regardless of milling time, as shown in Fig.1. The surface area of the U_3O_8 powder oxidized at 450℃ even decreased after milling. These results may suggest that the U_3O_8 powder was agglomerated during the milling. However, when the cyclic heat treating was applied, the BET surface area of both U_3O_8 powers increased significantly. Fig. 1 shows that the BET surface area could be increased up to more than 3 times by applying the cyclic heat treating.

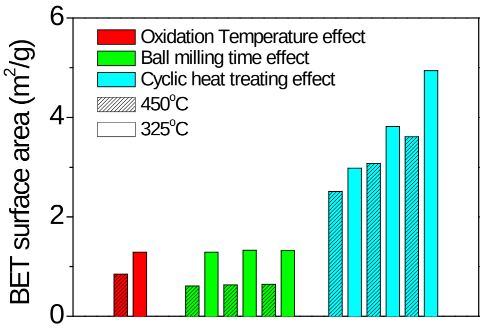

Fig. 1. BET surface area changes of the U_3O_8 powders.

The effect of prepared U_3O_8 powders on UO_2 pellet density has been tested. Fig. 2 shows the U_3O_8 added UO_2 pellet density changes according to the variation of milling time of U_3O_8 powder. The pellet density was slightly increased when the milled U_3O_8 powders were added in UO_2. Fig. 1 and Fig.2 indicate that density increase is responsible for the increase of BET surface area of U_3O_8 powder.

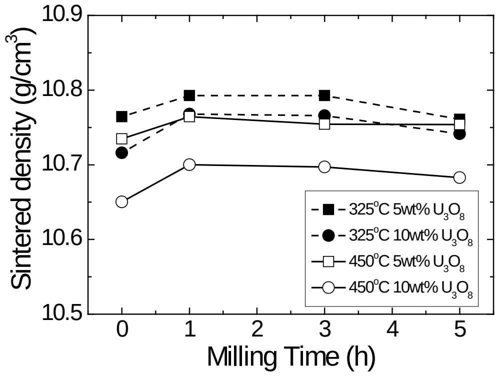

Fig. 2. Milling effect of recycled U_3O_8 powders on sintered UO_2 pellet density

Fig. 3 shows the density change of U_3O_8 added UO_2 pellets according to the repeating number of the cyclic heat treatment. In this curve, the pellet density is largely increased by the addition of the one cyclic heat treated U_3O_8 powder. However, pellet density is saturated or slightly decreased when two or three-cyclic heat treated powders are added. This result indicates that the certain range of BET surface area of U_3O_8 powder could be effective in mitigating the density drop of UO_2 pellets.

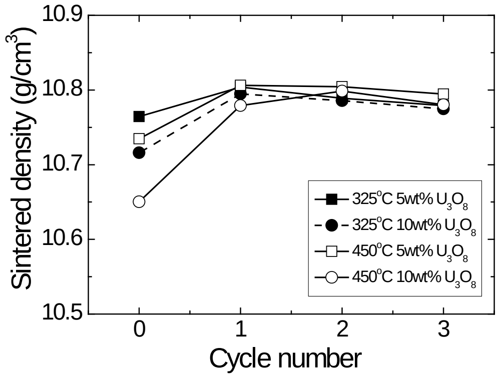

Fig. 3. Cyclic heat treatment effect of U_3O_8 powder on sintered UO_2 pellet density.

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REFERENCES

[1] OECD, NEA report No. 6224, Very High burn-ups in Light Water Reactors.

[2] K.W. Kang , J.H. Yang, J.H. Kim, Y.W. Rhee, D.J. Kim, K.S. Kim, K.W. Song, "Effects of MnO-Al2O3 on the Grain Growth and High Temperature Deformation Strain of UO_2 Fuel Pellets", Journal of Nuclear Science and Technology 47(2010) 304-307

[3] H. Assmann, H. Bairiot, in: Guidebook on quality Control of Water Reactor Fuel, Tech. Report Series No. 221. IAEA, Vienna, 1983